

the filler [includes a mixture of] comprising from about 20 to about 50 wt.% of a first filler and from about 50 to about 80 wt.% of a second filler, the first filler being selected from talc, mica, wollastonite, or combinations thereof, and the second filler being selected from calcium carbonate, barium sulfate, or combinations thereof.

### **REMARKS**

Claims 1-46 are pending in the application. Claims 23-46 have been cancelled without prejudice in response to the restriction requirement. Claims 1, 2, 15, 16, and 20 have been amended. No new matter has been added. Submitted herewith is a clean set of pending claims. Therefore, claims 1-22 remain in the application. Reconsideration of these claims in view of the following remarks is respectfully requested.

#### **Drawing**

To obviate the objection of the drawing, the Applicants are submitting under separate cover an amended drawing with proper margins. A copy of that submission is being included herewith. No new matter has been entered and it is respectfully requested that the objection be withdrawn.

#### **Election/Restriction**

The Applicants are confirming the election to prosecute claims 1-22. Claims 23-46 are being cancelled without prejudice to obviate the restriction requirement.

#### **35 U.S.C. § 112, First Paragraph, Rejection**

Claims 14 and 19 were rejected because it was stated that the specification fails to describe the "impact copolymer polypropylene" claimed or how to make it. To assist in showing that the term "impact copolymer polypropylene" is known in the art, the Applicants are submitting a declaration of one of the co-inventors, Wen P. Wu, under C.F.R. § 1.132 (Exhibit A). The term

“impact copolymer polypropylene” is a term that is known to one of ordinary skill in the art. See Wu Decl ¶ 5. Impact copolymer polypropylenes are physical mixtures of homopolymer polypropylene and random copolymer polypropylene. *Id.* and see, e.g., Exhibit B (“Handbook of Polypropylene and Polypropylene Composites” that describes various types of polypropylenes). Therefore, it is respectfully requested that this rejection be withdrawn.

### **35 U.S.C. § 103(a) Rejection**

#### Independent Claim 1

The Applicants submit that a *prima facie* case of obviousness under 35 U.S.C. § 103(a) over U.S. Patent No. 5,225,466 to Akao (“Akao”), the JP 61032743A abstract, or the combination thereof has not been made.

First, Akao does not teach or suggest, *inter alia*, a filler that includes “a high aspect ratio filler and a low aspect ratio filler, the high aspect ratio filler having an aspect ratio of at least about 5:1 and the low aspect ratio filler having an aspect ratio of less than about 3:1, the filler comprising at least about 50 wt.% of the low aspect ratio filler” as recited in claim 1.

Rather, Akao discloses “[t]he reinforcing material includes calcium carbonate, potassium titanate fiber, barium sulfate, magnesium carbonate, magnesium hydroxide, glass fiber, silica, titanium dioxide, alumina, talc, pyrophyllite, kaolin, sericite, mica, clay, etc. Col. 4, lines 22-26.

Akao also discloses “wherein said reinforcing material is a member selected from includes calcium carbonate, potassium titanate fiber, barium sulfate, magnesium carbonate, magnesium hydroxide, glass fiber, silica, titanium dioxide, alumina, talc, mica, clay, or a mixture thereof” Col. 10, lines 13-18. In all of the Examples, Akao only discloses the use of one single reinforcing material (15 wt.% talc). See col. 7, line 13 - col. 9, line 15. Thus, Akao does not teach or suggest combining

the claimed high aspect ratio filler and low aspect ratio filler, let alone in the recited amount of claim 1.

The JP 61032743A abstract, the other applied reference, does not address the deficiencies of Akao. Specifically, the JP 61032743A abstract does not teach or suggest, *inter alia*, a filler that includes "a high aspect ratio filler and a low aspect ratio filler, the high aspect ratio filler having an aspect ratio of at least about 5:1 and the low aspect ratio filler having an aspect ratio of less than about 3:1, the filler comprising at least about 50 wt.% of the low aspect ratio filler" as recited in claim 1. Rather, the JP 61032743A abstract discloses "[t]he inorganic filler is, e.g., [sic, e.g.,] mica or talc, having high aspect ratio and an average dia. of pref. 1.5-5 microns." (underlining in original). Therefore, it is believed that a *prima facie* case of obviousness under 35 U.S.C. § 103(a) over Akao, the JP 61032743A abstract, or the combination thereof has not been made.

Assuming, *arguendo*, that a prima facie case has been made, claim 1 should be allowable over Akao, JP 61032743A abstract, or the combination thereof because of the unexpected results of the claimed invention. To assist in showing the unexpected results obtained, a declaration of Wen P. Wu is being submitted herewith as Exhibit A. The declaration of Wen Wu discusses tests that were performed on several different mineral-filled polymeric sheets. *See generally* Wu Decl. at ¶¶ 6-10. These tests, including the procedures, were reported in Table 1 and discussed at pages 7-9 of the present application.

Inventive Examples 10-18 reported in Table 1 were talc-filled polypropylene sheets with ratios of high aspect ratio filler (HAR) to low aspect ratio filler (LAR) that ranged from 50:50 to 25:75. Table 1 at page 8 of the present application and Wu Decl. ¶ 6. In Inventive Examples 10-18, the tested properties of the mineral-filled polymeric sheets were generally improved as the

amount of filler increased with the ratio of the high aspect ratio filler to the low aspect ratio filler (HAR/LAR ratio) remaining constant. *See* Table 1 at page 8 of the present application and Wu Decl. ¶ 7.

It was surprising that at a constant HAR/LAR ratio in Inventive Examples 10-18 that the Gardner Impact values remained about the same or even increased in value when the amount of filler increased in the sheet. *See* Wu Decl. ¶¶ 6,7 discussing Table 1 with Gardner Impact values in Inventive Examples 10-12 (9.2, 9.7, 8.6 in-lbs), Inventive Examples 13-15 (11.0, 10.8, 12.3 in-lbs) and Inventive Examples 16-18 (13.6, 14.6, 21.2 in-lbs), and page 9, lines 16-18 of the present application. As shown in Table 1, the flexural modulus and tensile modulus also increased in Inventive Examples 10-18 when the filler amount was increased at a constant HAR/LAR ratio. Wu Decl. ¶¶ 6,7 discussing and comparing Inventive Examples 10-12, 13-15 and 16-18 from Table 1 of the present application. The mineral-filled polymeric sheets of Inventive Examples 10-18 had a desirable combination of properties (flexural modulus, tensile modulus and Gardner Impact). Wu. Decl. ¶ 8 and page 9, lines 18-19 of the present application.

The Comparative Examples 1-9 and 19-21, on the other hand, did not have a desirable combination of properties as compared to Inventive Examples 10-18. Wu Decl. ¶ 9. Specifically, the Gardner Impact values of Comparative Examples 1-9 were relatively small and decreased as the amount of filler increased at a constant HAR/LAR ratio. *See* Wu Decl. ¶ 6, 9 discussing Table 1 with Comparative Examples 1-3 (3.0, 1.6, 0.4 in-lbs), Comparative Examples 4-6 (5.8, 4.4, 3.3 in-lbs) and Comparative Examples 7-9 (5.9, 4.3, 2.4 in-lbs), and page 9, lines 3-4 of the present application. Comparative Examples 19-21 had desirable Gardner Impact values that increased as the amount of filler increased, but had lower flexural modulus and tensile modulus than is

generally desired. *See* Wu Decl. ¶ 6, 9 discussing Table 1, and page 9, lines 23-26 of the above-identified invention.

Thus, in summary, it was unexpected to one skilled in the art that the claimed combination and amount of the high aspect ratio filler and low aspect ratio filler in the polymeric sheet produced desirable combinations of properties.

Thus, independent claim 1 should not be obvious under 35 U.S.C. § 103(a) over Akao, the JP 61032743A abstract, or the combination thereof. Claims 2-15, which depend either directly or indirectly from claim 1, should be allowable for at least those reasons.

#### Independent Claims 16 and 20

Independent claim 16 recites a filler that comprises “from about 20 to about 50 wt.% talc and from about 50 to about 80 wt.% calcium carbonate.” Independent claim 20 recites a filler that comprises “from about 20 to about 50 wt.% of a first filler and from about 50 to about 80 wt.% of a second filler, the first filler being selected from talc, mica, wollastonite, or combinations thereof, and the second filler being selected from calcium carbonate, barium sulfate, or combinations thereof.” As discussed above with respect to claim 1, neither Akao, nor JP 61032743A abstract, nor the combination thereof teaches or suggests combining a high aspect ratio filler and low aspect ratio filler, let alone the specific combinations and amounts recited in respective claims 16 and 20.

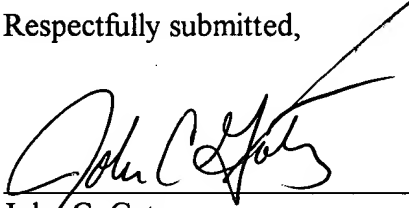
Thus, independent claims 16 and 20 should not be obvious under 35 U.S.C. § 103(a) over Akao, the JP 61032743A abstract, or the combination thereof. Claims 17-19 and 21-22, which depend either directly or indirectly from claim 16 or 20, should be allowable for at least those reasons.

**Conclusion**

It is the Applicants' belief that all of the claims are now in condition for allowance, and action towards that effect is respectfully requested.

If there are any matters which may be resolved or clarified through a telephone interview, the Examiner is requested to contact the undersigned attorney at the number indicated.

Respectfully submitted,

A handwritten signature in black ink, appearing to read "John C. Gatz", is written over a horizontal line.

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John C. Gatz  
Reg. No. 41,774  
Jenkins & Gilchrist  
225 West Washington Street, Suite 2600  
Chicago, IL 60606-3418  
(312) 425-3900

Attorney for Applicants

**Clean Set Of Pending Claims**  
**U.S. Patent Application No. 09/850,985**

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1. (Once Amended) A container comprising a base having a bottom wall and a continuous base wall, the continuous base wall encompassing the bottom wall and extending upwardly therefrom, the base comprising a polyolefin and a filler, the base comprising from about 30 to about 75 wt.% filler and from about 25 to about 70 wt.% polyolefin, the filler including a high aspect ratio filler and a low aspect ratio filler, the high aspect ratio filler having an aspect ratio of at least about 5:1 and the low aspect ratio filler having an aspect ratio of less than about 3:1, the filler comprising at least 50 wt.% of the low aspect ratio filler.
  2. (Once Amended) The container of claim 1, wherein the container further comprises a lid, the lid having a top wall and a continuous lid wall, the lid wall encompassing the top wall and extending downwardly therefrom, the lid comprising a polyolefin and a filler, the base comprising from about 30 to about 75 wt.% filler and from about 25 to about 70 wt.% polyolefin, the filler including a high aspect ratio filler and a low aspect ratio filler, the high aspect ratio filler having an aspect ratio of at least about 5:1 and the low aspect ratio filler having an aspect ratio of less than about 3:1, the filler comprising at least 50 wt.% of the low aspect ratio filler.
  3. The container of claim 1, wherein the high aspect ratio filler has an aspect ratio of from about 5:1 to about 40:1.
  4. The container of claim 3, wherein the high aspect ratio filler has an aspect ratio of from about 10:1 to about 20:1.
  5. The container of claim 1, wherein the low aspect ratio filler has an aspect ratio of from 1:1 to about 2:1.
  6. The container of claim 1, wherein the filler comprises from about 50 to about 80 wt.% low aspect ratio filler and from about 20 to about 50 wt.% high aspect ratio filler.

7. The container of claim 1, wherein the high aspect ratio filler is talc, mica, wollastonite, or combinations thereof.

8. The container of claim 7, wherein the high aspect ratio filler is talc.

9. The container of claim 1, wherein the low aspect ratio filler is calcium carbonate, barium sulfate, or the combination thereof.

10. The container of claim 9, wherein the low aspect ratio filler is calcium carbonate.

11. The container of claim 1, wherein the polyolefin is a polypropylene, a polyethylene, or combinations thereof.

12. The container of claim 11, wherein the polyolefin is a polypropylene.

13. The container of claim 12, wherein the polyolefin is a polypropylene homopolymer.

14. The container of claim 12, wherein the polyolefin is an impact copolymer polypropylene.

✓  
A 15. (Once Amended) The container of claim 1, wherein the base comprises from about 35 to about 65 wt.% filler and from about 35 to about 65 wt.% polyolefin.

16. (Once Amended) A container comprising a base having a bottom wall and a continuous base wall, the continuous base wall encompassing the bottom wall and extending upwardly therefrom, the base comprising a polyolefin and a filler, the filler comprising from about 20 to about 50 wt.% talc and from about 50 to about 80 wt.% calcium carbonate.

17. The container of claim 16, wherein the polyolefin is a polypropylene, a polyethylene, or combinations thereof.



18. The container of claim 17, wherein the polyolefin is a homopolymer polypropylene.

19. The container of claim 17, wherein the polyolefin is an impact copolymer polypropylene.

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AB 20. (Once Amended) A container comprising a base having a bottom wall and a continuous base wall, the continuous base wall encompassing the bottom wall and extending upwardly therefrom, the base comprising a polymer and a filler, the filler comprising from about 20 to about 50 wt.% of a first filler and from about 50 to about 80 wt.% of a second filler, the first filler being selected from talc, mica, wollastonite, or combinations thereof, and the second filler being selected from calcium carbonate, barium sulfate, or combinations thereof.

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21. The container according to claim 20, wherein the first filler is talc.

22. The container according to claim 20, wherein the second filler is calcium carbonate.

49. Photonic Polymer Systems: Fundamentals, Methods, and Applications, edited by Donald L. Wise, Gary E. Wnek, Debra J. Trantolo, Thomas M. Cooper, and Joseph D. Giesse

50. Handbook of Polymer Testing: Physical Methods, edited by Roger Brown

51. Handbook of Polypropylene and Polypropylene Composites, edited by Harutun G. Karian

#### Additional Volumes in Preparation

Polymer Blends and Alloys, edited by Gabriel O. Shonaike and George P. Simon

Star and Hyperbranched Polymers, edited by Munmaya K. Mishra and Shiro Kobayashi

Practical Extrusion Blow Molding, edited by Samuel L. Belcher

Polymer Viscoelasticity: Stress and Strain in Practice, Everisto Riancho, Ricardo Diaz-Calleja, Catalina Salom, Margalit Prolongo, and Rosa Mesegosa

# HANDBOOK OF POLYPROPYLENE AND POLYPROPYLENE COMPOSITES

edited by

**HARUTUN G. KARIAN**

*Thermofil, Inc.  
Brighton, Michigan*



MARCEL DEKKER, INC.

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It has outstanding retention of mechanical properties after recycling (see also in many cases incineration ability). The competitive price comes from the availability of the basic monomer and the simplicity of the polymerization available today. The polymer can be processed on a wide variety of equipment, thus allowing it to be transformed easily, simply, and safely into a wide variety of usable articles. For these reasons, polypropylene has a bright future as one of humankind's building materials for the next century.

## 2

# Polypropylene: Structure, Properties, Manufacturing Processes, and Applications

William J. Kissel and James H. Han

*Amoco Polymers, Inc., Alpharetta, Georgia*

Jeffrey A. Meyer

*Amoco Corporation, Naperville, Illinois*

## 2.1 TYPES OF POLYPROPYLENE

Polypropylene (PP) is a thermoplastic material that is produced by polymerizing propylene molecules, which are the monomer units, into very long polymer molecules or chains. There are a number of different ways to link the monomers together, but PP as a commercially used material in its most widely used form is made with catalysts that produce crystallizable polymer chains. These give rise to a product that is a semicrystalline solid with good physical, mechanical, and thermal properties. Another form of PP, produced in much lower volumes as a byproduct of semicrystalline PP production and having very poor mechanical and thermal properties, is a soft, tacky material used in adhesives, sealants, and caulk products. The above two products are often referred to as "isotactic" (crystallizable) PP (i-PP) and "atactic" (noncrystallizable) PP (a-PP), respectively.

As is typical with most thermoplastic materials, the main properties of PP in the melt state are derived from the average length of the polymer chains and the breadth of the distribution of the polymer chain lengths in a given product. In the solid state, the main properties of the PP material reflect the type and amount of crystalline and amorphous regions formed from the polymer chains.

Semicrystalline PP is a thermoplastic material containing both crystalline and amorphous phases. The relative amount of each phase depends on structural and stereochemical characteristics of the polymer chains and the conditions under

which the resin is converted into final products such as fibers, films, and various other geometric shapes during fabrication by extrusion, thermoforming, or molding.

Polypropylene has excellent and desirable physical, mechanical, and thermal properties when it is used in room-temperature applications. It is relatively stiff and has a high melting point, low density, and relatively good resistance to impact. These properties can be varied in a relatively simple manner by altering the chain regularity (tacticity) content and distribution, the average chain lengths, the incorporation of a comonomer such as ethylene into the polymer chains, and the incorporation of an impact modifier into the resin formulation.

The following notation is used in this chapter: Polypropylene containing only propylene monomer in the semicrystalline solid form is referred to as homopolymer PP (HPP), and we use this to mean the i-PP form. Polypropylene containing ethylene as a comonomer in the PP chains at levels in about the 1–8% range is referred to as random copolymer (RCP). HPP containing a comonomer RCP phase that has an ethylene content of 45–65% is referred to as an impact copolymer (ICP). Each of these product types is described below in more detail.

### 2.1.1 Homopolymer

Homopolymer PP is the most widely used polypropylene material in the HPP, RCP, and ICP family of products. It is made in several different reactor designs using catalysts that link the monomers together in a stereospecific manner, resulting in polymer chains that are crystallizable. Whether they crystallize and to what extent depends on the conditions under which the entangled mass of polymer chains transitions from the melt to the solid state or how a heat-softened solid PP material is strained during a further fabrication procedure like fiber drawing.

Homopolymer PP is a two-phase system because it contains both crystalline and noncrystalline regions. The noncrystalline, or amorphous, regions are comprised of both isotactic PP and atactic PP. The isotactic PP in the amorphous regions is crystallizable, and it will crystallize slowly over time up to the limit that entanglement will allow. The extent of crystallization after the initial fabrication step of converting PP pellets or powder into a molded article will slowly increase over time, as will the stiffness. A widely accepted model of HPP morphology likens the solid structure to a system consisting of pieces of stiff cardboard linked together by strands of softer material. In the areas represented by flat pieces of cardboard, PP polymer chains weave up and down into close-packed arrays called crystallites ("little crystals"), which are called lamella by morphologists. The soft strands linking the pieces of stiff cardboard are polymer chains that exit one crystallite, enter another, and then begin weaving up and down in another crystallite. The crystallizability of the chains is one factor that determines how thick the crystallites will be and the thickness of the crystallites determines

## Characteristics of Polypropylene

how much heat energy is required to melt them (the melting temperature). A typical HPP has an array of crystallinities from thick ones to very thin ones, and these manifest themselves as an array of melting points.

Homopolymer PP is marketed mainly by melt flow rate (MFR) and additive formulation into fiber, film, sheet, and injection molding applications. Melt flow rate is an indicator of the weight-average molecular weight as measured by the ASTM or ISO MFR test method.

### 2.1.2 Random Copolymer

Random copolymers are ethylene/propylene copolymers that are made in a reactor by copolymerizing propylene and small amounts of ethylene (usually and lower). The copolymerized ethylene changes the properties of the polymer chains significantly and results in thermoplastic products that are sold into markets in which slightly better impact properties, improved clarity, decreased haze, decreased melting point, or enhanced flexibility are required. The ethylene monomer in the PP chain manifests itself as a defect in the chain regularity, thus inhibiting the chain's crystallizability. As the ethylene content increases, the crystallinity thickness gradually decreases, and this manifests itself in a lower melting point. The amount of ethylene incorporated into the chain is usually dictated by a balance between the thermal, optical, or mechanical properties.

### 2.1.3 Impact Copolymers

Impact copolymers are physical mixtures of HPP and RCP, with the overall mixture having ethylene contents on the order of 6–15% w/w. These are sold into markets where enhanced impact resistance is needed at low temperatures, especially freezer temperature and below.

The RCP part of the mixture is designed to have ethylene contents on the order of 40–65% ethylene and is termed the rubber phase. The rubber phase can be mechanically blended into the ICP by mixing rubber and HPP in an extruder or it can be polymerized in situ in a two-reactor system. The HPP is made in the first reactor and the HPP with active catalyst still in it is conveyed into a second reactor where a mixture of ethylene and propylene monomer is polymerized. The voids and interstices of the HPP polymer powder particle. The amount of rubber phase that is blended into the HPP by mechanical or reactor methods determined by the level of impact resistance needed. The impact resistance of the ICP product is determined not only by its rubber content but also by the size, shape, and distribution of the rubber particles throughout the ICP product. Reactor product is usually give better impact resistance at a given rubber level for the reason.

As the rubber content of the ICP product is increased, so is the impact resistance.

tance, but this is at the expense of the stiffness (flexural modulus) of the product. Consequently, polymer scientists often describe a product as having a certain impact-stiffness balance. The stiffness of the ICP product is dictated by the stiffness of the HPP phase and the volume of rubber at a given rubber size distribution in the product. The impact resistance is dictated by the amount and distribution of the rubber phase in the ICP product.

## 2.2 TACTICITY

The solid-state characteristics of PP occur because the propylene monomer is asymmetrical in shape. It differs from the ethylene monomer in that it has a methyl group attached to one of the olefinic carbons. This asymmetric nature of the propylene monomer thus creates several possibilities for linking them together into polymer chains that are not possible with the symmetrical ethylene monomer, and gives rise to what are known as structural isomers and stereochemical isomers in the polypropylene chain.

In structural isomerism, polymer scientists refer to the olefinic carbon with the methyl group on it as the "head" (h) and the other olefinic carbon as the "tail" (t) of the monomer. The most common method of polymerization uses catalysts that link the monomers together in the "head-to-tail" fashion, although occasionally there is a "mistake" made and the monomers form a "head-to-head" or a "tail-to-tail" linkage, but these tend to be rare.

Stereochemical isomerism is possible in PP because propylene monomers can link together such that the methyl groups can be situated in one spatial arrangement or another in the polymer. If the methyl groups are all on one side of the chain, they are referred to as being in the "isotactic" arrangement, and if they are on alternate sides of the chain, they are referred to as being in the "syndiotactic" arrangement. Each chain has a regular and repeating symmetrical arrangement of methyl groups that form different unit cell crystal types in the solid state. A random arrangement of methyl groups along the chain provides little or no symmetry, and a polymer with this type of arrangement is known as "atactic" polypropylene.

When polymer scientists discuss the stereochemical features of PP, they usually discuss it in terms of "tacticity" or "percent tacticity" of polypropylene, and in the marketplace the term "polypropylene" is generally used to refer to a material that has high tacticity, meaning high isotactic content. The high tacticity PP materials have desirable physical, mechanical, and thermal properties in the solid state. Atactic material is a soft, sticky, gummy material that is mainly used in sealants, caulks, and other applications where its stickiness is desirable. Syndiotactic PP, not a large volume commercial material, is far less crystalline than isotactic PP.

## Characteristics of Polypropylene

### 2.3 MOLECULAR WEIGHT AND MOLECULAR WEIGHT DISTRIBUTION

Unlike pure simple compounds, whose molecules are all of the same molecular weight, polymer samples consist of molecules of different molecular weight. This is a reflection of the fact that a polymer sample is a collection of molecules of differing chain lengths. Therefore, an average molecular weight concept was adopted for polymers. No single average, however, can completely describe a polymer sample, and a number of different averages are used. Ratios of some of these averages can be used to calculate a molecular weight distribution (MWD) which describes the breadth of the molecular weights represented.

Molecular weight averages of polypropylene are measured by the technique of gel permeation chromatography (GPC), a chromatographic technique that separates out the polymer chains by chain length after the PP is dissolved in a solvent. When dissolved, the PP is no longer a thermoplastic but instead is a bunch of long molecules dispersed in a solvent. From the GPC data, one can calculate the number-average ( $M_n$ ), weight-average ( $M_w$ ), and z-average ( $M_z$ ) molecular weights. In PP, the  $M_z$  relates to physical properties of the solid, the  $M_w$  relates to viscosity properties of the melt, and the  $M_n$  to elastic properties of the melt. Because the GPC chromatogram contains a lot of data that is not easy to tabulate and communicate, it is convenient to use a ratio, especially  $M_w/M_n$ , because it gives a good estimate of the MWD and is a simple number to tabulate and store. It is a good estimate because the  $M_n$  is very sensitive to short chains and the  $M_w$  is very sensitive to long chains in the products.

The GPC equipment is fairly expensive and prone to failure, and the actual experiment is slow, labor intensive, and requires dissolution of the PP at high temperatures in solvents like xylene and trichlorobenzene. Thus, other methods to estimate the molecular weight have been developed. The most popular method is termed the MFR test, and it gives a number that is easily correlatable to average. Most HPP products are sold with MFR numbers ranging from 0.2 to 45, and these correspond to  $M_w$ s from 1,000,000 down to 100,000. Note that molecular weight averages are inversely proportional to MFR numbers.

### 2.4 MECHANICAL PROPERTIES

The mechanical properties of most interest to the PP product design engineer are its stiffness, strength, and impact resistance. Stiffness is measured as the flexural modulus, determined in a flexural test, and impact resistance by a number of different impact tests, with the historical favorite being the Izod impact at ambient and at subambient temperatures. These mechanical properties are mostly used to predict the properties of molded articles. Strength is usually defined by the stress-strain curve.